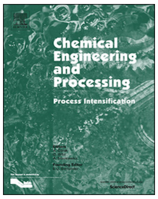
Chemical Engineering & Processing: Process Intensification 132 (2018) 1–10

Contents lists available at ScienceDirect

Chemical Engineering & Processing: Process Intensification

journal homepage: www.elsevier.com/locate/cep



Adsorption-catalytic process for removal of volatile organic compounds from lean waste gases: Optimization of the adsorbent-catalyst bed geometry

S. Zazhigalov[a,b], N. Chumakova[a,b], A. Zagoruiko[a,b,*]

[a] Boreskov Institute of Catalysis, Ave. Lavrentieva, 5, Novosibirsk, 630090, Russia
[b] Novosibirsk State University, Pirogova St., 2, Novosibirsk, 630090, Russia

ARTICLE INFO

Keywords:
Adsorption-catalytic process
Volatile organic compounds
Abatement
Mathematical modeling
Optimal bed shape

ABTSTRACT

The study was dedicated to the adsorbent-catalyst bed geometry optimization in the adsorption-catalytic process for the removal of volatile organic compounds (VOC) by means of the mathematical modeling. The commercial software COMSOL Multiphysics was used for the 2D axisymmetric modeling of the process with internal location of the heater, which initiates the self-sufficient adsorbent-catalyst regeneration. The model took into account the adsorption and oxidation reactions on the internal pellet surface, mass transfer between gas flow and catalyst bed, diffusion in the pellets, and heat transfer in the bed. The simulation showed the failure of adsorbent-catalyst regeneration in the cylindrical bed. At the same time, the beds with inlet part in a shape of truncated cone appeared to be much more efficient. The different cone side angles and inflow gas rates were considered by means of mathematical modeling. The optimal geometry parameters for the best process performance were defined in the study.

1. Introduction

The industrial waste gases purification from volatile organic compounds (VOCs) is an actual problem in atmospheric air protection. Such impurities (hydrocarbons, alcohols, acids, esters, aldehydes, etc.) are present in waste gases of various industries, such as chemicals and petrochemicals, machinery, wood processing, printing, and many others.

Catalytic oxidation of VOCs into harmless products (CO2 and water) is the most feasible technology for processing of waste gases with moderate and low VOC content. Most catalytic reactions of VOC deep oxidation occur at elevated temperatures. This temperature depends upon the VOC specifics and on the catalyst type, but for majority of practical applications it equals to at least 200–300 °C. At the same time, the described waste gases typically have low (ambient) temperature, so the catalytic combustion processes require some additional energy for heating of such gases to reaction temperatures. Therefore, the energy consumption is one of the most important parameters, defining the selection of abatement technology.

The steady-state catalytic combustion processes may be performed without input of external energy for gases with relatively high VOC content (above 4–5 g/m^3). The more lean gases may be neutralized by means of the catalytic reverse-flow process [1,2] providing the autothermal gas purification for gases with moderate VOC content (down to ~1 g/m^3). The treatment of waste and vent gases with lower VOC concentration, often accompanied with high volume of gases to be purified, is the real-life problem as it requires high energy consumption and capital cost of purification units.

The combining of adsorption and catalytic technologies in adsorption-catalytic processes is of a special interest in the solution of this problem. Such adsorption-catalytic processes are based on adsorption of VOCs directly on the catalyst surface at ambient temperature with periodic regeneration of the catalyst by oxidation of the adsorbed impurities at the same catalyst at elevated temperatures [3–13]. The process performance includes periodic alteration of adsorption and regeneration cycles. The gas to be purified enters the adsorbent-catalyst bed at the adsorption cycle with ambient temperature, without any preheating. When the VOCs appear in the outlet gases, the adsorption cycle switches to the catalyst regeneration cycle. At this process cycle, the inlet gas is heated by the heater situated upstream the reactor entrance. Heating of the catalyst initiates the oxidation of adsorbed VOCs, creating the heat front of oxidation reaction in the catalyst, slowly moving along the direction of the gas flow. When the regeneration is finished, the adsorbent-catalyst is ready again for the next adsorption cycle.

This process allows to avoid the permanent heating of the feed gas thus giving the way to significantly reduce the energy consumption during the treatment of low-concentrated gases, compared to

---

[*] Corresponding author at: Boreskov Institute of Catalysis, Ave. Lavrentieva, 5, Novosibirsk, 630090, Russia.
E-mail address: zagor@catalysis.ru (A. Zagoruiko).

https://doi.org/10.1016/j.cep.2018.08.002
Received 20 March 2018; Received in revised form 24 July 2018; Accepted 4 August 2018

Nomenclature

[A]	Product of reversible adsorption
[]	Oxidized site at the catalyst surface
[P]	Product of irreversible chemisorption on internal pellet surface
A	VOC molecule
a	Catalyst adsorption capacity, wt. %
c_i	Gas flow component (VOC or oxygen) concentration, mol/m^3
$c_{c,i}$	Gas component (VOC or oxygen) concentration in the pellet pores, mol/m^3
$C_{P,c}$ C_P	Catalyst and gas heat capacity, J/kg/K
C_s	Maximum surface VOC concentration, mol/m^2
$D_{c,i}$ $D_{e,i}$	Effective diffusion coefficients, m^2/s
$D_{F,A}$ $D_{F,O}$	VOC and oxygen diffusion coefficients, m^2/s
$h_{D,i}$	Mass transfer coefficients m/s
K	Bed permeability, m^2
M_{VOC}	VOC molar mass, g/mol
N	Number of pellets per unit volume of the bed
Q	Heat source, W/m^3
Q_i	Thermal effects, kJ/m^3
r	Pellet radial coordinate, dimensionless
r_c	Pellet radius, m
R_c	Reaction source term, mol/m^3/s
S_i	Mass source term, mol/m^3/s
S_b	Reactive specific surface area, m^2/m^3
S_s	Catalyst specific surface area, m^2/g
u	Gas velocity vector, m/s
y	Distance from the reactor wall, m
α	Cone angle, °
ε_p,ε_c	Bed and catalyst porosity
λ_c, λ	Thermal conductivity of the catalyst and gas, W/m/K
μ	Gas dynamic viscosity, kg/m/s
ρ	Gas density, kg/m^3
ρ_c	Catalyst density, kg/m^3

conventional abatement technologies. Moreover, the significant energy saving may be provided in the new version of the adsorption-catalytic process [14], where the regeneration is initiated by the heater located directly inside the catalytic bed. Such approach allows to reduce the energy consumption for the regeneration initiation by at least two orders of magnitude compared to the conventional version of the process (Fig. 1).

In the case of the external heater location (Fig. 1a) all the inlet gas should be heated up to the VOCs ignition temperature (~300–350 °C). Accounting for heat losses for heating of inlet pipe and reaction chamber, it may take up to 5 min of continuous heating for the regeneration to start and occur in autothermal regime, finally resulting in high energy consumption. If the regeneration initiator is situated inside the bed (Fig. 1b) we have to heat relatively small (~1 kg) amount of catalyst up to the ignition temperature, thus reducing the energy consumption, especially in the case of the large inlet gas volumes (> 1000 m^3/h).

The regeneration efficiency and energy consumption in the adsorption-catalytic process obviously depends upon many factors, such as: chosen catalyst, gas flow rate, temperature of gas to be purified, heater power, and others. In addition, the shape of the adsorbent-catalyst bed, defining the propagation of the heat wave at the regeneration stage, may also play an important role. In our previous studies, we used the 1D model, which provides the adequate description of adsorption and reaction phenomena in the axial bed of pseudo-cylindrical geometry with diameter of embedded heater equal to the diameter of catalyst bed. At the same time, such model is inapplicable for simulation of these phenomena in the cases of other bed shapes and transverse size of heated area is smaller compared to bed diameter.

The aim of this study was optimization of the adsorbent-catalyst bed shape by means of the 2D axisymmetric modeling, reproducing the 3D geometry of the different axisymmetric adsorbent-catalyst beds.

2. Mathematical model of the adsorption-catalytic process

2.1. Model description

The 2D axisymmetric model was considered to describe the adsorption-catalytic process in the reactors of different shape. The modelling was performed using the COMSOL Multiphysics commercial software.

The chemistry of the processes was described by simplified kinetic scheme [15] that includes the reversible VOCs adsorption (1), the transition of physically adsorbed VOCs ($[A]$) into the irreversibly adsorbed chemisorbed form ($[P]$) (2), and the oxidation of the chemisorption intermediates into deep oxidation products (3):

$$A + [\,] \leftrightarrow [A] \tag{1}$$

$$[A] \rightarrow [P] \tag{2}$$

$$[P] + \nu\, O_2 \rightarrow CO_2 + H_2O + [\,] \tag{3}$$

The in-built physical interfaces were used for the model equations assignment. The catalytic bed was presented as porous medium with the “Reactive Pellet Bed” interface that gives an extra-dimension related to the pellet radius. So the material balance in the bed was set by the “Transport of Diluted Species in Porous Media” interface (4) with included “Reactive Pellet Bed” module (5).

$$\varepsilon_p \frac{\partial c_i}{\partial t} - \nabla\bullet(D_{e,i}\nabla c_i) + u\bullet\nabla c_i = S_i \tag{4}$$

$$4\pi N\left\{r^2 r_c^2 \varepsilon_c \frac{\partial c_{c,i}}{\partial t} - \nabla\bullet(r^2 D_{c,i}\nabla c_{c,i}) = r^2 r_c^2 R_{c,i}\right\} \tag{5}$$

where the effective diffusion coefficients are $D_{c,i} = D_{e,i} = \varepsilon_p^{4/3} D_{F,i}$.

The flux of mass across the pellet-gas interface is set as

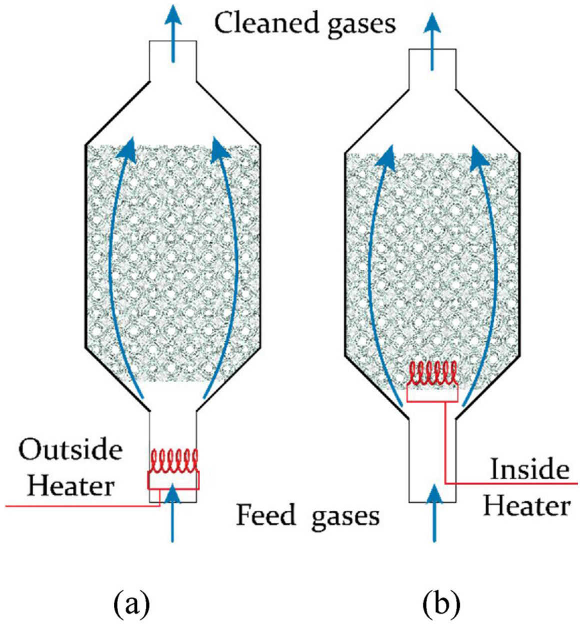


Fig. 1. Flow sheets of the conventional adsorption-catalytic process (a) and the process with the internal heater disposition (b).